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TITLE MICROWAVE SINTERING OF ALUMINA-SiC COMPOSITES AT 2.45 AND 60 GHz

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Abstract

Composites of alumina-10v/o silicon carbide whiskers were sintered to 70% of theoretical density using both 2.45 and 30 GHz microwave radiation. Further densification was inhibited by formation of a constrained network of whiskers. Alumina-10v/o silicon carbide platelet composites were sintered to 94% of theoretical density with 2.45 GHz microwaves in less than 20 minutes.

Introduction

Microwave processing is a suitable technique for a wide range of ceramic materials. Among the ceramics which can be successfully heated and sintered using microwaves are, alumina, zirconia, boron carbide and alumina-silicon carbide composites. Several advantages over conventional heating techniques are realized because energy is coupled directly to the ceramic body. First, since heating is volumetric instead of by conduction from the surface to the interior, a more uniform microstructure and consequently, better mechanical properties result. Thermal stresses upon heat-up are also minimized because of the homogeneous nature of volumetric heating. Minimization of heat-up stresses will ultimately allow the fabrication of more complex parts and shapes than are possible by conventional processing. Additionally, processing times are much shorter and energy costs lower. The longest processing time for any of the samples reported in this paper was 20 minutes. To date microwave sintering studies to produce 95% dense boron carbide have resulted in an 18% energy savings over conventional induction heated hot-pressing.

Heating results from the energy dissipated by a ceramic placed in a microwave field. The power dissipated by a dielectric in an electric field is given by the following equation:

$$P = (2\pi f \epsilon)(E^2/2) \tan \delta$$

where P is the power dissipation, f is the frequency, ϵ is the dielectric permittivity, E is the electric field strength and tan δ is the loss tangent. It is readily apparent from this equation, that in order for significant heating to occur the ceramic must have an appreciable loss tangent and dielectric permittivity.

Experimental

Sumitomo alumina, grade AKP-50*, was used for the matrix material. The manufacturer quotes a mean particle size of 0.2 microns and a B.E.T. specific surface area of 11 m^2/gm. Vapor-solid type silicon carbide whiskers manufactured by, Huber** were used for the whisker reinforcement material. The manufacturer specifies the material to be beta silicon carbide with a diameter from 0.3-1 microns and a length of 5-50 microns. The platelets used in this study were obtained from American Matrix⁺. The platelets are alpha silicon carbide, 10-10,000 microns in diameter and 1-10 microns thick.

The alumina-whisker mixture was placed in a Bel-Art⁺⁺ Micro-Mill and blended for two minutes. The platelets and alumina were mixed for one hour in a glass baffle jar on a rolling mill. These mixing procedures were followed by uniaxial pressing (without binders or sintering aids) to 10,000 psi and then by isostatic pressing to 50,000 psi.

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*Sumitomo Chemical Company, Ltd., Osaka, Japan.
**J.M. Huber Inc., Borger, TX.
⁺American Matrix Inc., Knoxville, TN.
⁺⁺Bel-Art Products, Pequannock, NJ.

These blending and pressing procedures resulted in 1 cm diameter X 1 cm high pellets of approximately 55% green density for the alumina-whisker composite and approximately 60% green density for the alumina-platelet composite.

2.45 GHz microwave processing was performed in-house at Los Alamos National Laboratory. The LANL facility, shown in Fig. 1, consists of a 6 kilowatt microwave power supply, a 2 ft X 2 ft X 2 ft resonant cavity equipped with a rotating specimen table and a two-color infrared pyrometer. 60 GHz microwave processing was performed at Varian Associates in Palo Alto, CA. The 60 GHz experimental setup is shown in Fig. 2. This setup consists of a high voltage power supply (which is not visible since it is under the floor), a gyrotron tube which is encased in the blue steel-lead-lined box, a flow-through reaction chamber and a two-color infrared pyrometer. A wave guide extends from the gyrotron to the platform on the mezzanine level where the reaction chamber and two color infrared pyrometer are setup. The reaction chamber is shown in Fig. 3. Also visible in this photograph is an alumina foam sample holder containing two green samples.

All specimens were heated from room temperature to the peak temperature. Upon achieving the peak temperature, power was shut-off and the sample allowed to cool to room temperature.

Results and Discussion

Figure 4 is a plot of percent theoretical density verses peak sintering temperature for the alumina-10 v/o silicon carbide whisker composites processed using 2.45 GHz microwave radiation. The density appears to be independent of temperature between 1500 and 1840°C, varying between 68 and 72% in a random manner.

Independence of density with sintering temperature over such a large range is certainly unusual, but can be readily explained by examination of the microstructure. A typical as sintered microstructure is shown in Fig. 5. Examination of this photomicrograph reveals that crack-like voids have opened up as a result of densification. The cause of crack formation appears to be shrinkage of the alumina matrix away from a constraining whisker network during densification. Since there are only 10 v/o whiskers in the composite the observation of sintering inhibition due to formation of a constraining network is unexpected.

Based on the work of Onoda[1], a constraining network of inert particles is usually thought to occur only after the inert particles occupy 20 v/o or more of the green compact. Recently Lange[2] has proposed a constrained network model for predicting the densification behavior of composites. He has suggested, based on percolation theory, that much smaller volume fractions of an inert phase could form many short continuous paths which could act as a constrained network.

Figure 6 is a plot of percent theoretical density versus peak sintering temperature for alumina-10 v/o silicon carbide whiskers processed under 60 GHz microwave radiation. Because of limited access to the high frequency apparatus, only a limited number of specimens were run. For these samples the sintering temperature ranged from 1240 to 1620°C. Between 1450 and 1620°C, the density varied from 68 to 70%, which is similar to the results achieved for samples processed using 2.45 GHz microwaves. So, for similar sintering temperatures both 2.45 and 60 GHz microwave radiation resulted in similar densities. Meek et al.[3] in an earlier study of 60 GHz microwave sintering of Sumitomo alumina-10 v/o Arco* silicon carbide whiskers reported a density of 77% for the sintering temperatures of 1450°C and 1740°C. This density is somewhat higher than the densities reported here, and is thought to result from the use of different silicon carbide whiskers with a smaller aspect ratio. Tiegs and Becher[4] have previously shown that whisker aspect ratio can have a large effect on composite densification. These workers have also reported a density of 71% for the conventional pressureless sintering of Bailkowski** alumina-10 v/o Arco silicon carbide whiskers at a temperature between 1700 and 1800°C. The difference in density reported by Meek et al.[3] and Tiegs and Becher[4] is probably due to differences in green compaction and aluminas.

*Arco Chemical Co., Greer, SC.
**Baikowski Int'l Corp., Charlotte, NC.

As might be expected from the density results, the microstructures of samples sintered using either microwave frequency are very similar. Figure 7 is a side-by-side comparison of a sample processed at 2.45 GHz and one processed at 60 GHz. The microstructures both exhibit void-like cracking and whisker clumps which are artifacts of the dry blending process. Whisker clumps such as these can be expected to result in poor mechanical properties. Clearly, a more sophisticated blending procedure is necessary to produce ceramic composites suitable for engineering applications. In spite of these scattered clumps the whiskers are otherwise randomly and uniformly distributed within the alumina matrix as shown in Fig. 8. It should also be pointed out that the development of cracks upon densification in no way appears to be related to the whisker clumps.

Figure 9 is a plot of percent theoretical density versus peak sintering temperature for alumina-10^v/o silicon carbide platelets. For sintering temperatures in the range of 1620 to 1650°C, densities of 94% are typical. Figure 10 is a photomicrograph of a 94% dense alumina-10^v/o silicon carbide platelet composite. The platelets appear to be uniformly distributed with the exception of a few agglomerates. A typical agglomerate is shown at higher magnification in Fig. 11. A white phase, which has not yet been identified, is associated with the agglomerates.

It is interesting to consider these results in terms of a constraining network model. For both the whisker and platelet composites 10^{v}/o of the reinforcement phase was added to the same matrix. A constrained network resulted for the whisker composite but not for the platelet composite. These results clearly show that the aspect ratio of the inert phase has a very large effect on the formation of a constrained network in a composite.

Conclusions

For alumina-10^{v}/o silicon carbide whisker composites both 2.45 and 60 GHz microwave processing yielded similar densities at similar sintering temperatures. A limiting density of about 70% resulted due to the formation of a constrained whisker network which caused void-like cracks to open upon densification of the matrix phase. The same type of void-like cracking has been reported by Lange[2] for the conventional pressureless sintering of a similar composite. The overriding factor in the densification of this composite is not the method of heating, whether it be conventional or microwave, but the formation of a constraining network.

Alumina-10^{v}/o silicon carbide platelets have been sintered to 94% of theoretical density using 2.45 GHz microwaves. Sintering was performed without the use of sintering aides or binders and in under 20 minutes.

It has been shown that alumina-10^{v}/o silicon carbide composites can be readily heated and sintered using 2.45 GHz microwaves in minutes. The problems encountered in densifing alumina-10v/o sillicon carbide whisker composites are typical of that system and not characteristic of microwave processing.

Acknowledgment
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References

1.) G.Y.Onoda, Jr.,"in *Ceramic Microstructures '76*", edited by R.M. Fulrath and J.A.Pask, (Westview, Boulder, CO. 1977), pp. 163-183.

2.) F.F.Lange,"*Constrained Network Model for Predicting Densification Behavior of Composite Powders*". J. Mater. Res., **2**(1), pp. 59-65, 1987.

3.) T.T.Meek, R.D.Blake and J.J.Petrovic,"*Microwave Sintering of Al_2O_3 and Al_2O_3-SiC Whisker Composites*", Ceram. Eng. Sci. Proc., **8**(7-8), pp. 861-871, 1987.

4.) T.N.Tiegs and P.F.Becher, "*Sintered Al_2O_3-SiC Whisker Composites*", Am. Ceram. Soc. Bull., **66**(2), pp. [illegible], 1987.

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1.) Photograph of Los Alamos 2.45 GHz microwave processing facility. The power supply is on the left, the microwave cavity is on the right and the two color IR pyrometer is in the foreground.

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9.) Plot of percent theoretical density verses maximum sintering temperature for alumina-10 v/o silicon carbide platelets processed at 2.45 GHz. Densities as high as 94 % of theoretical were obtained by heating to 1640°C.

10.) Photomicrograph of 94 % dense alumina-10 v/o silicon carbide platelets. The platelets are uniformly distributed except for a few agglomerates.

11.) Photomicrograph of a platelet agglomerate. Unidentified white phase appears to be associated with [illegible].

Figure 2

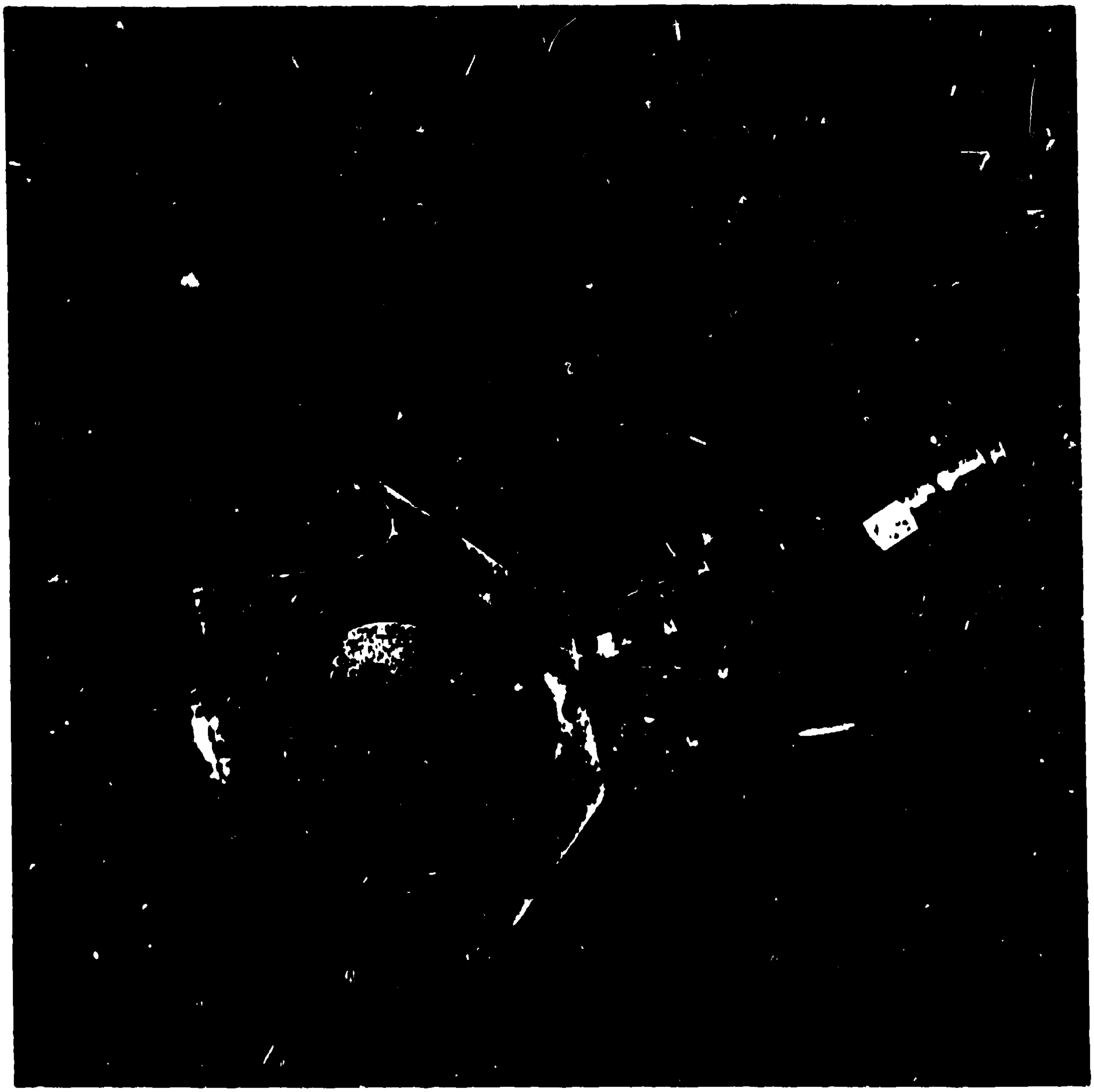
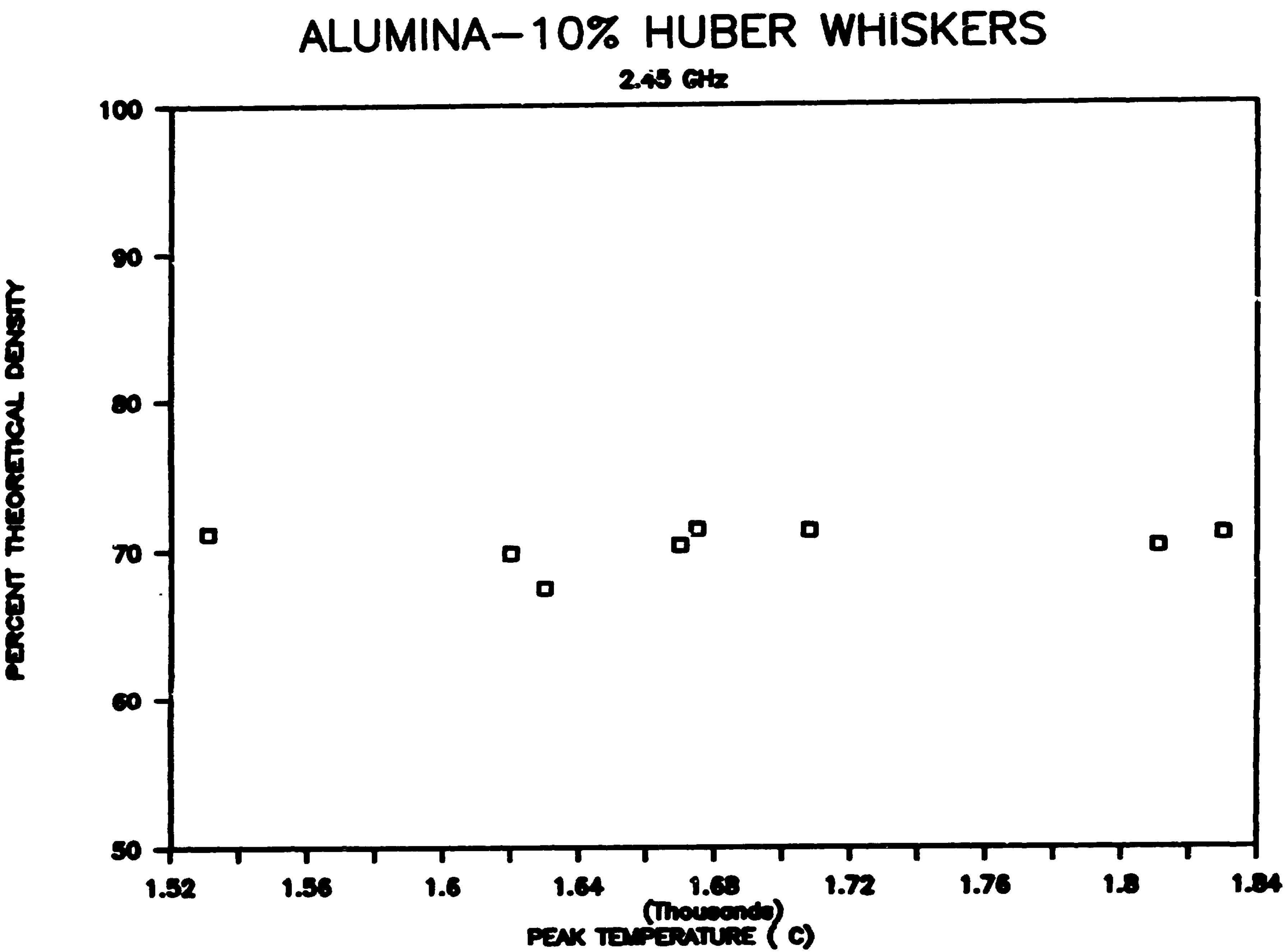


Figure 3



ALUMINA-10% HUBER WHISKERS
2.45 GHz
PERCENT THEORETICAL DENSITY
100
90
80
70
60
50
1.52
1.56
1.6
1.64
1.68
1.72
1.76
1.8
1.84
(Thousands)
PEAK TEMPERATURE (C)

Figure 4

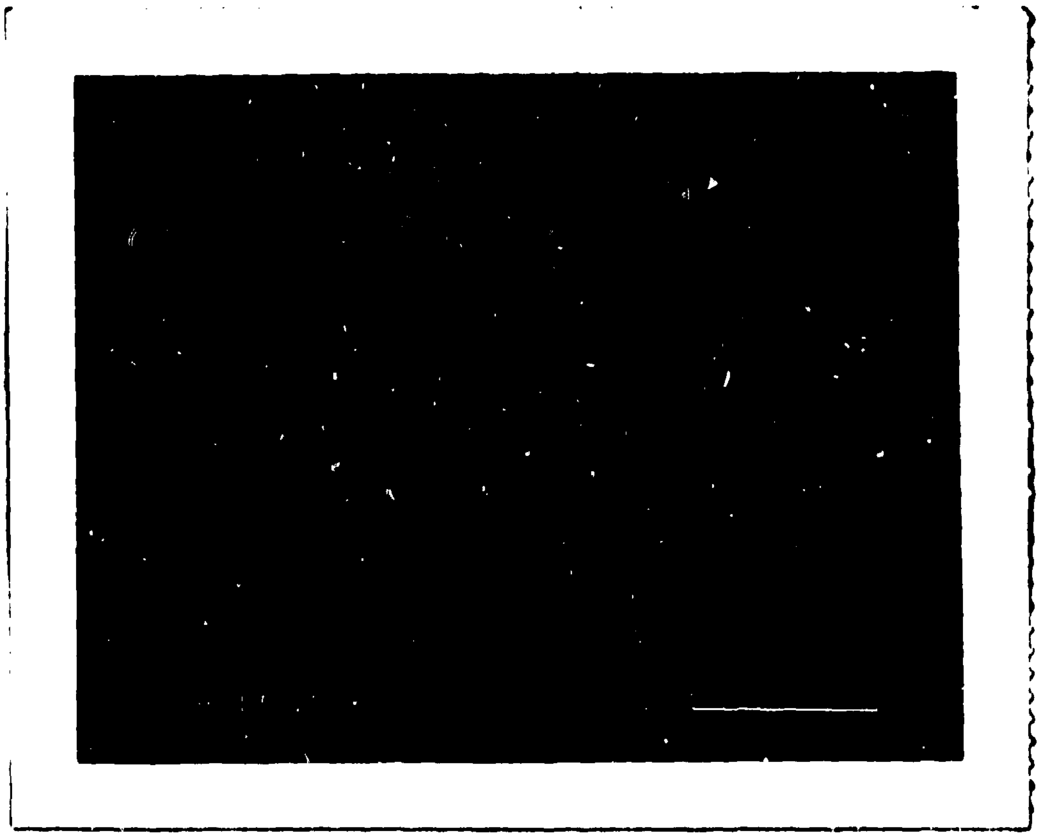
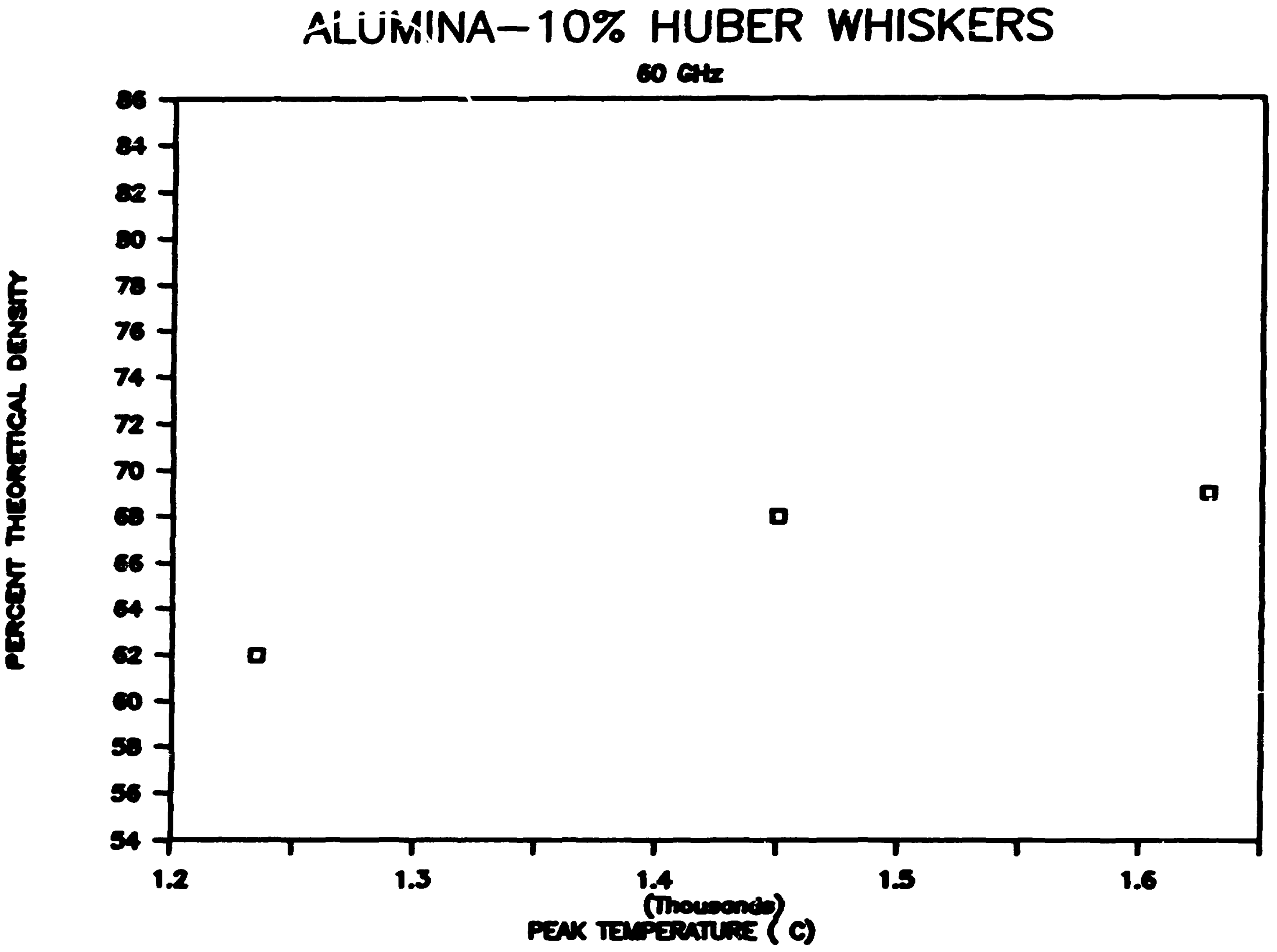
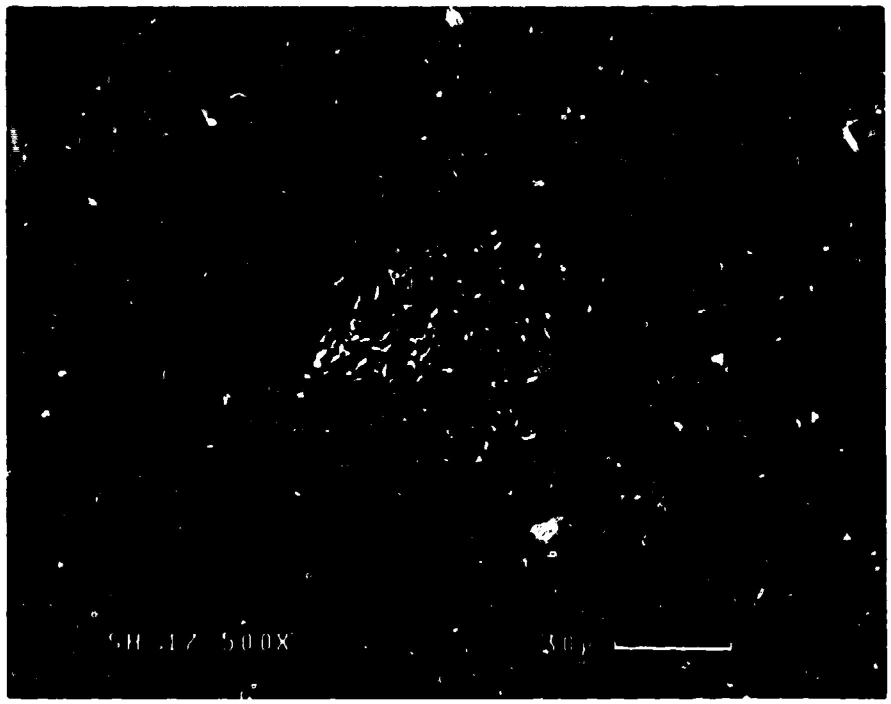


Figure 5


ALUMINA–10% HUBER WHISKERS
60 GHz
PERCENT THEORETICAL DENSITY
86
84
82
80
78
76
74
72
70
68
66
64
62
60
58
56
54
1.2
1.3
1.4
1.5
1.6
(Thousands)
PEAK TEMPERATURE (C)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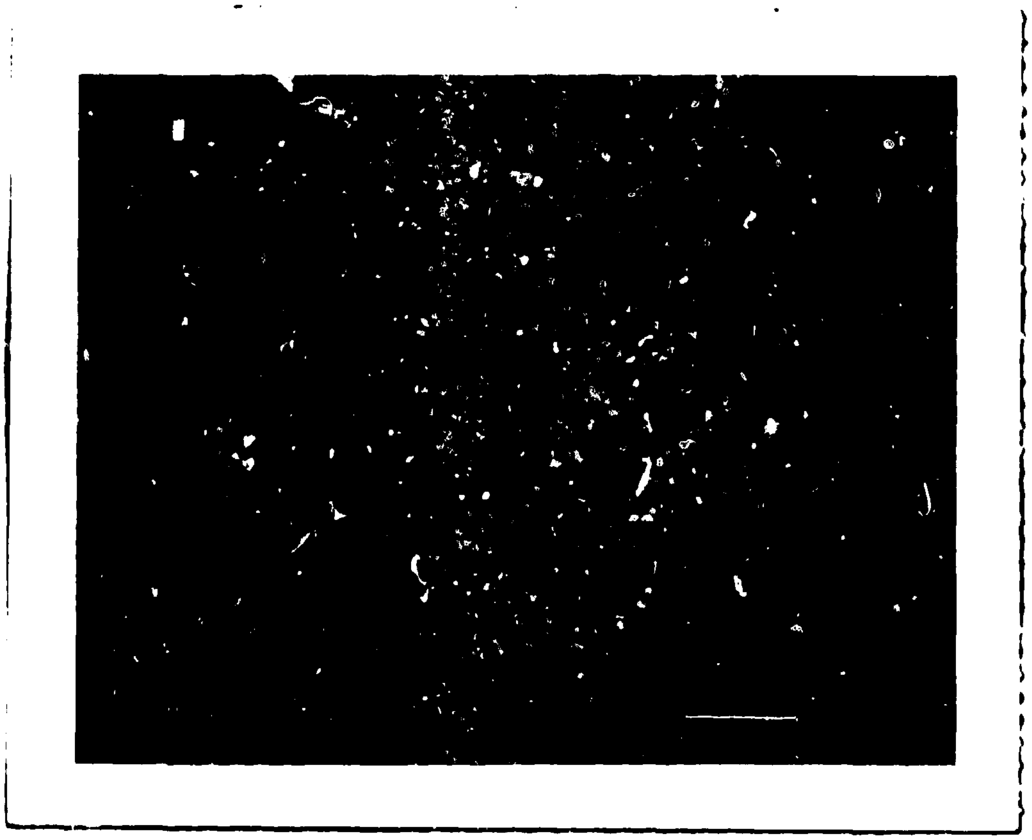
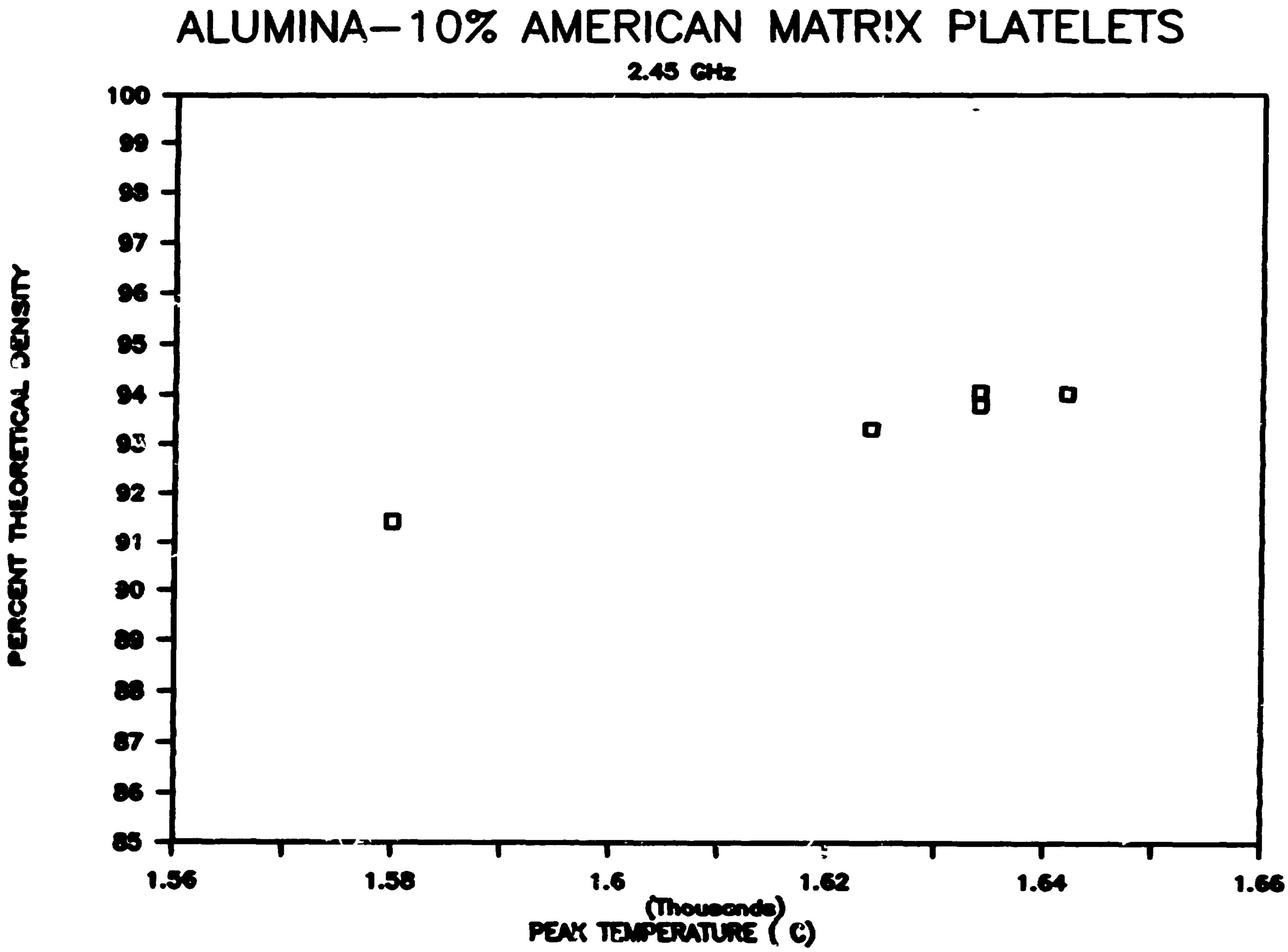
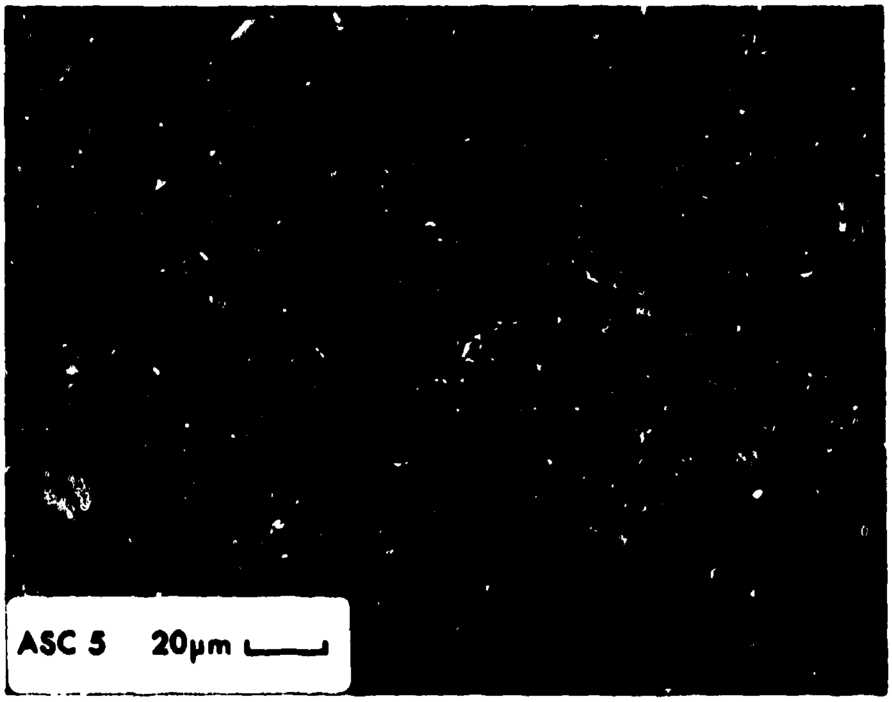
30μ





ALUMINA-10% AMERICAN MATR!X PLATELETS
2.45 GHz
PERCENT THEORETICAL DENSITY
100
99
98
97
96
95
94
93
92
91
90
89
88
87
86
85
1.56
1.58
1.6
1.62
1.64
1.66
(Thousands)
PEAK TEMPERATURE (C)


ASC 5
20µm

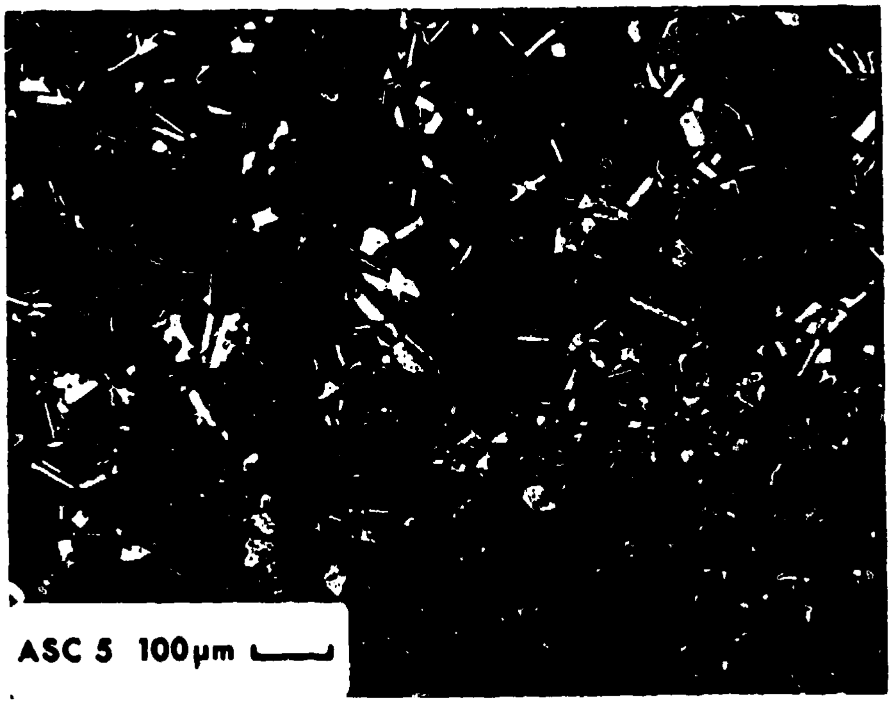


Figure 11